Review

Behaviour and fate of perfluoroalkyl and polyfluoroalkyl substances (PFASs) in drinking water treatment: A review

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ABSTRACT

This article reviews perfluoroalkyl and polyfluoroalkyl substance (PFAS) characteristics, their occurrence in surface water, and their fate in drinking water treatment processes. PFASs have been detected globally in the aquatic environment including drinking water at trace concentrations and due, in part, to their persistence in human tissue some are being investigated for regulation. They are aliphatic compounds containing saturated carbon–fluorine bonds and are resistant to chemical, physical, and biological degradation. Functional groups, carbon chain length, and hydrophilicity/hydrophobicity are some of the important structural properties of PFASs that affect their fate during drinking water treatment. Full-scale drinking water treatment plant occurrence data indicate that PFASs, if present in raw water, are not substantially removed by most drinking water treatment processes including coagulation, flocculation, sedimentation, filtration, biofiltration, oxidation (chlorination, ozonation, AOPs), UV irradiation, and low pressure membranes. Early observations suggest that activated carbon adsorption, ion exchange, and high pressure membrane filtration may be effective in controlling these contaminants. However, branched isomers and the increasingly used shorter chain PFAS replacement products may be problematic as it pertains to the accurate assessment of PFAS behaviour through drinking water treatment processes since only limited information is available for these PFASs.

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Contents

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Nomenclature

AOP	advanced oxidation process
CCL3	3rd contaminant candidate list
DI	de-ionized
DOC	dissolved organic carbon
FASAs	perfluoroalkane sulfonamides
FOSA	perfluorooctance sulfonamide
FASEs	perfluoroalkane sulfonamidoethanols
GAC	granular activated carbon
IX	ion exchange
MIEX	magnetic ion exchange resin
MW	molecular weight
MWCO	molecular weight cut-off
N-EtFOSE	N-ethyl perfluorooctane-sulfonamido ethanol
N-MeFOSE	N-methyl perfluorooctane-sulfonamido ethanol
NF	nanofiltration
NOM	natural organic matter
O_3	ozone
OH	hydroxyl radical
PAC	powdered activated carbon
PFAAs	perfluoroalkyl acids

PFASs	perfluoroalkyl and polyfluoroalkyl substances
PFBA	perfluorobutanoic acid
PFBS	perfluorobutane sulfonic acid
PFCAs	perfluoroalkyl carboxylic acids
PFDA	perfluorodecanoic acid
PFDoDA	perfluorododecanoic acid
PFHpA	perfluoroheptanoic acid
PFHxA	perfluorohexanoic acid
PFHxS	perfluorohexane sulfonic acid
PFNA	perfluorononanoic acid
PFOA	perfluorooctanoic acid
PFOS	perfluorooctane sulfonic acid
PFPnA	perfluoropentanoic acid
PFSAs	perfluoralkyl sulfonic acids
PFUnDA	perfluoroundecanoic acid
RO	reverse osmosis
4:2 FTOH	4:2 fluorotelomer alcohol
6:2 FTOH	6:2 fluorotelomer alcohol
8:2 FTOH	8:2 fluorotelomer alcohol
10:2 FTOH	10:2 fluorotelomer alcohol
USEPA	United States environmental protection agency
UV	ultraviolet
WWTP	wastewater treatment plant

1. Introduction

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are a diverse class of chemicals that have in common an aliphatic carbon backbone in which hydrogen atoms have been completely (prefix: per-) or partially (prefix: poly-) replaced by fluorine. These substances, owing to their highly polar and strong carbon–fluorine bonds, have some unique chemical attributes including extremely high thermal and chemical stability. They are primarily used as surfactants in numerous industrial and consumer products such as firefighting foams, alkaline cleaners, paints, non-stick cookware, carpets, upholstery, shampoos, floor polishes, fume suppressants, semiconductors, photographic films, pesticide formulations,

food packaging, masking tape, denture cleaners, etc. (e.g. Kissa, 2001; Brooke et al., 2004).

This review follows the terminology recommended by Buck et al. (2011) and uses PFAS instead of the more commonly used acronym PFC (perfluorinated compound). PFASs are characterized by their functional groups. Table 1 presents the structures and some important environmental properties of selected, most prominently studied PFASs. There are numerous other PFAS compounds in use, for example phosphorus containing PFASs which have only very recently been detected in surface, drinking, and waste waters (D'Eon et al., 2009; Ding et al., 2012). Further details regarding structure and nomenclature of PFASs are provided in Buck et al. (2011). Those that to-date have received most attention are the

perfluoroalkyl acids (PFAAs), perfluoroalkyl sulfonamides (FASAs), and telomer alcohols (FTOHs). Two important classes of PFAAs are the perfluoroalkyl carboxylic acids (PFCAs) and the perfluoroalkyl sulfonic acids (PFSAs). A wide range of perfluoroalkyl chain lengths and branching patterns exists.
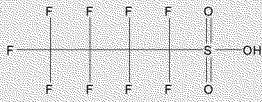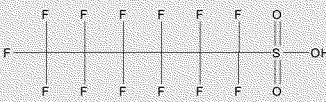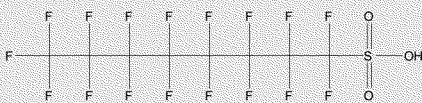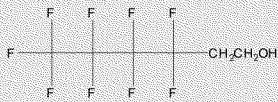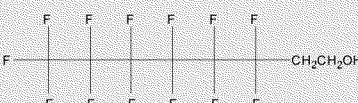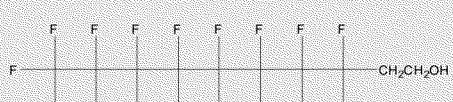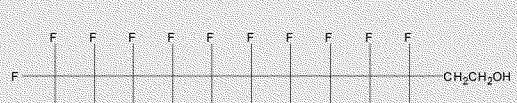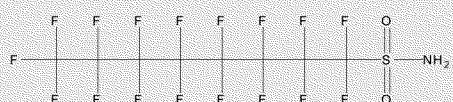
Since PFASs are usually applied in technical mixtures both linear and branched isomers occur in the environment. However, the current lack of knowledge about the detailed composition of these technical mixtures and the inaccessibility of suitable analytical standards for branched isomers

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Table 1 — Structure and physico-chemical properties of selected perfluoroalkyl and polyfluoroalkyl substances (PFASs).

Compound name & CAS registry#	Structure	MW[a]	log K_{OC} (L/kg)	Solubility (mg/L)	Vapour pressure (Pa)
Perfluoroalkyl acids (PFAAs)					
Perfluoroalkyl carboxylic acids (PFCAs)					
Perfluorobutanoic acid (PFBA) [375-22-4]		214.1			851[b] (25 C)
Perfluoropentanoic acid (PFPeA) [2706-90-3]		264.1			
Perfluorohexanoic acid (PFHxA) [307-24-4]		314.1			
Perfluoroheptanoic acid (PFHpA) [375-85-9]		364.1		118,000[c] (21.6 C)	20.89[b] (25 C)
Perfluorooctanoic acid (PFOA) [335-67-1]		414.1	1.47[d]	4340[c] (24.1 C)	4.17[b] (25 C)
Perfluorononanoic acid (PFNA) [375-95-1]		464.1	2.06[d]		1.29[b] (25 C)
Perfluorodecanoic acid (PFDA) [335-76-2]		514.1	2.37[d]	260[c] (22.4 C)	0.23[b] (25 C)
Perfluoroundecanoic acid (PFUnDA) [2058-94-8]		564.1	2.32[d]	92.3[c] (22.9 C)	0.10[b] (25 C)
Perfluorododecanoic acid (PFDoA) [307-55-1]		614.1			0.008[b] (25 C)
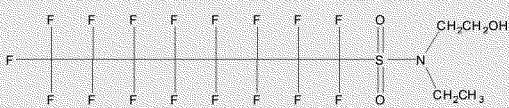
Table 1 — (continued)

Compound name & CAS registry#	Structure	MW[a]	log K_{OC} (L/kg)	Solubility (mg/L)	Vapour pressure (Pa)
Perfluoroalkyl sulfonic acids (PFSAs)					
Perfluorobutane sulfonic acid (PFBS) [375-73-5]		300.1		510[e]	
Perfluorohexane sulfonic acid (PFHxS) [355-46-4]		400.1	0.97[d]		
Perfluorooctane sulfonic acid (PFOS) [1763-23-1]		500.1	2.10[d]	570[f]	3.31 10⁻⁴ (25 C)[f]
Precursor compounds — fluorotelomer alcohols, perfluoroalkane sulfonamides and perfluoroalkane sulfonamidoethanols					
Fluorotelomer alcohols (FTOHs)					
4:2 Fluorotelomer alcohol (4:2 FTOH) [2043-47-2]		264.1	0.93[g]	974[g] (22.5 C)	992[h] (25 C)
6:2 Fluorotelomer alcohol (6:2 FTOH) [647-42-7]		364.1	2.43[g]	18.8[g] (22.5 C)	713[h] (25 C)
8:2 Fluorotelomer alcohol (8:2 FTOH) [678-39-7]		464.1	3.84[i]	0.194[i] (22.3 C)	254[h] (25 C)
10:2 Fluorotelomer alcohol (10:2 FTOH) [865-86-1]		564.1	6.20[g]	0.011[g]	144[h] (25 C)
Perfluoroalkane sulfonamides (FASAs)					
Perfluorooctane sulfonamide (FOSA) [754-91-6]		499.14	2.56[d]		
N-Alkyl Perfluoroalkane sulfonamidoethanols (FASEs)					
N-methyl perfluorooctane sulfonamidoethanol (N-MeFOSE) [24448-09-7]		557.22		0.81[k] (25 C)	0. 70[j] (25 C)

Table 1 — (continued)

Compound name & CAS registry#	Structure	MW[a]	log K_{OC} (L/kg)	Solubility (mg/L)	Vapour pressure (Pa)
N-ethyl perfluorooctane sulfonamidoethanol (N-EtFOSE) [1691-99-2]		571.25		0.89[b] (25 C)	0.35[i] (25 C)

Data presented in this table are mostly experimental data; detailed model predicted data can be found at Bhhatarai and Gramatica (2011).
[a] United States National Library of Medicine (2011).
[b] Bhhatarai and Gramatica (2011).
[c] Kaiser et al. (2006).
[d] Awad et al. (2011).
[e] Jensen et al. (2008).
[f] Stock et al. (2009).
[g] Liu and Lee (2007).
[h] Stock et al. (2004).
[i] Liu and Lee (2005).
[j] Lei et al. (2004).

make it challenging to quantify many PFAS isomers accurately in environmental matrices. This constrains the understanding of the fate and toxicity of individual PFAS isomers in the environment, and also limits our understanding of their behaviour in water treatment processes.

Most PFASs are extremely resistant to degradation (e.g. Kissa, 2001) and have therefore been detected ubiquitously in the aquatic environment. Some have even been detected at low concentrations in drinking water (pg/L to μg/L) making it a potential PFAS exposure route for humans. Post et al. (2012) reviewed available information on PFOA, its sources and occurrence in drinking water, toxicokinetics, and health effects. Information covered in their review "suggests that the continued human exposure to even low concentrations of PFOA in drinking water results in elevated body burdens that may increase the risk of health effects."

Earlier reviews (Rayne and Forest, 2009; Vecitis et al., 2009; Lutze et al., 2011; Eschauzier et al., 2011) on removal of PFASs from drinking water and wastewater focused primarily on bench-scale studies and have discussed various conventional and promising, though less commonly employed treatment options (e.g. photolysis, sonolysis, thermolysis etc.). However, there is a growing body of literature on PFAS in full-scale water treatment plants. Thus, the objective of this article is to critically review and summarize published PFAS drinking water treatment data reported in full-scale plants and to explain, where possible, the underlying mechanisms for the observed behaviour of PFASs by integrating the findings of select bench-scale studies. To provide further context this review also includes brief summaries of the occurrence of PFASs in source water, their toxicological significance and regulatory status, occurrence of PFASs in drinking water globally, and PFAS properties relevant to drinking water treatment.

1.1. Occurrence in the aquatic environment

Giesy and Kannan (2001) were among the first to report the widespread distribution of PFASs, which are released in the environment during their industrial production and

application, and also as a result of leaching from, and degradation of, consumer products. Eventually, PFASs enter wastewater treatment plants (WWTPs) and as such WWTPs have been suggested as one of the major point sources of PFASs to surface waters (Boulanger et al., 2005; Sinclair and Kannan, 2006; Moeller et al., 2010; Xiao et al., 2012a) and the atmosphere (Ahrens et al., 2011). In addition, discharge of PFASs contained in industrial waste or biosolids has been reported to contaminate surface and groundwater (Paustenbach et al., 2007; Hölzer et al., 2008; Minnesota Department of Health, 2008). Degradation of compounds such as FTOHs and FASAs leads to the formation of PFAAs (Ellis et al., 2003; Dinglasan et al., 2004; Wallington et al., 2006; Stock et al., 2007) and hence, these are often termed PFAA precursors.

High water solubility, simultaneous hydrophobic/hydrophilic properties, and low volatility of most PFAA contribute to their presence in all aquatic environments and even rain water. Although about 40 different PFASs have been detected in water (e.g. Ahrens, 2011), most studies have targeted PFOS and PFOA since, in many cases where several PFASs were monitored in water, PFOS and PFOA were detected more frequently and at the highest concentrations (Yamashita et al., 2005; Hoehn et al., 2007; Quinones and Snyder, 2009; Thompson et al., 2011a). Other frequently detected compounds include PFBA, PFHxA, PFHpA, PFNA, PFDA, PFBA, PFHxS, and FOSA (Table 1). PFBS and PFBA, two possible short chain replacement compounds for PFOS and PFOA (Renner, 2006; USEPA, 2012) were found to be the dominant PFASs in recent studies (Minnesota Department of Health, 2008; Moeller et al., 2010; Ahrens et al., 2010). As the regulations around PFOA and PFOS become more stringent it is probable that the use of other fluorinated organics will increase. In addition, many other PFASs not covered in this review are currently in use. Examples are phosphorus containing fluorinated organics such as polyfluoroalkyl phosphates (PAPs), perfluorinated phosphonic acids (PFPA), and perfluorinated phosphinic acids (PFPIA) which have been detected in surface water, wastewater, effluents and in drinking water (D'Eon et al., 2009; Ding et al., 2012). Hence, compounds other than

PFOA and PFOS should also be considered for monitoring studies.

Typical PFAS concentrations in water range from pg/L to ng/L. However, higher concentrations (μg/L to even mg/L) have been detected in surface and groundwater following fire-fighting activities or explosions (Moody and Field, 1999; Moody et al., 2002, 2003; Rumsby et al., 2009), and in some waters adjacent to fluorochemical manufacturing facilities (Hansen et al., 2002; Minnesota Department of Health, 2008; Hoffman et al., 2011). A critical review of the occurrence of PFASs in the aquatic environment has been published by Ahrens (2011). The occurrence of PFASs in drinking water is discussed in detail in Section 3.

1.2. Occurrence in humans

Low-level (typically ng/mL) concentrations of PFASs, notably PFOA and PFOS, has been detected in human tissue and blood serum worldwide (Kannan et al., 2004; Karrman et al., 2007; Monroy et al., 2008; Pan et al., 2010; Llorca et al., 2010; Ingelido et al., 2010; Liu et al., 2011). PFOA was detected in blood serum at mean concentrations of 122   81 and 424   333 ng/mL in two communities in Ohio that were exposed to PFOA-contaminated drinking water (Bartell et al., 2010). Emmett et al. (2006) have previously shown that drinking water contaminated with PFOA (released from the nearby DuPont Washington Water Works) was the major exposure route and the "residential water source was the primary determinant of serum PFOA."

1.3. Toxicity and regulatory framework

Although there is a growing body of literature on PFAS toxicity in animal models, data on the toxicological effects of PFASs on humans are limited (e.g. Steenland et al., 2010). Even for PFOA, "to-date data are insufficient to draw firm conclusions regarding the role of PFOA for any of the diseases of concern" (Steenland et al., 2010). However, a recently published study conducted on a large cohort of mid-Ohio valley residents that were exposed to contaminated drinking water or had worked at the local DuPont Washington Works chemical plant found PFOA to be associated with kidney and testicular cancer in that community (Barry et al., 2013). Other epidemiological studies have suggested a link between blood serum levels of certain PFASs and low birth weight (Fei et al., 2007), infertility-measured as longer waiting time to pregnancy (Fei et al., 2009), onset of early menopause in women (Knox et al., 2011), increased impulsivity and delayed puberty in children (Gump et al., 2011; Lopez-Espinosa et al., 2011), low semen quality in young men (Joensen et al., 2009), and thyroid disease in the US general adult population (Melzer et al., 2010). PFOA has recently been included on a list of 'obesogens', chemicals that may contribute to obesity (Janesick and Blumberg, 2011; Holtcamp, 2012). Longer chain carbon PFASs (>C8) have been reported to bioaccumulate in wildlife and humans (Hekster et al., 2003; Martin et al., 2003; Houde et al., 2008; Conder et al., 2008). Once PFASs enter the body they are poorly eliminated. The reported serum half-life of pefluorohexane sulfonate (PFHxS), PFOS, and PFOA in humans is 8.5 years, 5.4 years, and 2.9–8.5 years, respectively (USEPA, 2009; Seals et al., 2011) (Table SI-1 in supporting information). The slow elimination rates of PFASs suggest that "continued exposure could increase body burdens to levels that would result in adverse outcomes" (USEPA, 2009).

The USEPA has recently included PFOA and PFOS in its pared-down third drinking water contaminant candidate list (CCL3) of 32 compounds for further regulatory studies (USEPA, 2011a). The agency also included six PFASs (PFBS, PFHxS, PFOS, PFHpA, PFOA and PFNA) in its final list of 32 contaminants for the unregulated contaminants monitoring rule 3 (UCMR3) (USEPA, 2011b) thereby collecting occurrence data to assist with the development of future regulations should they be required. Drinking water advisory levels/goals/guideline values for PFOS and PFOA in various jurisdictions are listed in Table 2. It is evident that wide variations in drinking water guidelines among jurisdictions exist. This is likely due to differences in interpreting toxicity data or the safety factors taken into consideration to calculate those guideline values.

PFOS was recently listed as a persistent organic pollutant by the Persistent Organic Pollutants Review Committee (POPRC) of the United Nations Stockholm Convention on Persistent Organic Pollutants (POPRC, 2009; Wang et al., 2009) and efforts are underway in various jurisdictions in the developed world to limit or ban PFAS use (EU Directive, 2006; Government of Canada, 2008). A review of existing regulatory guidelines surrounding PFASs can be found in Zushi et al. (2012). However, concern for potential environmental release remains, in part due to emissions from the existing inventories. Also, while production in the US, Europe, and other developed countries becomes increasingly regulated, production of PFASs such as PFOS has been increasing sharply in other regions (USEPA, 2009) thereby merely shifting production from one region to another (Lindstrom et al., 2011). Hence, strong concerted global regulatory initiatives are highly desirable to address PFAS emissions on a global scale (Lindstrom et al., 2011).

2. PFAS properties

In fluorinated surfactants (including PFASs), the hydrophobic part of the molecule is either partially or completely fluorinated and can be straight chained or branched. The C–F bond is one of the strongest known and the bond is stronger with increasing replacement of hydrogen by fluorine at each carbon (O'Hagan, 2008). As such the more substituted the PFASs are, the less reactive (i.e. more chemically inert) they become. PFASs in general can withstand heat, acids, bases, reducing agents, oxidants, as well as photolytic, microbial, and metabolic degradation processes (Kissa, 2001; Schultz et al., 2003). Limited experimental data on hydrophobicity, acidity constants (pKa), and partitioning constants are available (Rayne and Forest, 2009) and what is available is often limited to linear forms of PFASs. The available experimental data and calculated pKa values indicate that both PFCAs and PFASs are strong acids which will predominantly be in their dissociated, negatively-charged form at environmentally relevant pH values (Kaiser et al., 2006; Rayne and Forest, 2009; Buck et al., 2011). Precursor compounds (i.e. FTOHs and FASAs) are generally neutral and will remain undissociated at pH values typically encountered in water.

Table 2 — Drinking water advisory levels/goals/guideline values for PFOA and PFOS.				
Regulatory body (jurisdiction)	PFOS (ng/L)		PFOA (ng/L)	References
USEPA (US) *Provisional health advisory value*	200		400	USEPA (2011c)
Minnesota Department of Health (MDH) (Minnesota, US)[a] *Health risk limit*	300		300	Minnesota Department of Health (2011)
New Jersey Department of Environmental Protection (New Jersey, US) *Health-based drinking water concentration for PFOA*			40	Post et al. (2009)
German Drinking Water Commission (Germany) *Health-based precautionary values*	Immediate precautionary action value (combined PFOA and PFOS) Infants and pregnant women: 500 Adult: 5000 Chronic precautionary action value (combined PFOA and PFOS): >100–600 ng/L; combined PFOA and PFOS value for maximum of 10 years >600–1500 ng/L for a maximum of 3 years			Trinkwasserkommission (2006)
Drinking water inspectorate (DWI) (UK) *Guidance values*	Tier 2	>300 Action: Monitor levels and consult with health professionals	>300	DWI (2009)
	Tier 3	>1000 Action: In addition to Tier 2 actions take measures to reduce concentration to <1000 ng/L and <10,000 ng/L for PFOS and PFOA, respectively as soon as is practicable.	>5000	
	Tier 4	>9000 Action: In addition to Tier 3 actions take measures to reduce exposure from drinking water within 7 days; ensure consultation with health professionals takes place as soon as possible.	>45,000	

[a] MDH has also set health guideline values for PFBS and PFBA at 7000 ng/L.

PFCAs and PFSAs have low vapour pressures which decrease with increasing carbon chain length. This suggests low potential for volatilization (Prevedouros et al., 2006) and hence, they are unlikely to be removed from drinking water by air stripping. FTOHs, FASAs and perfluoroalkane sulfonamidoethanols (FASEs) such as 8:2 FTOHs are much more volatile (indicated by relatively higher vapour pressure) than PFAAs (Table 1).

Water solubility of PFASs increases as carbon chain length decreases (Bhhatarai and Gramatica, 2011). PFCAs and PFSAs which carry a charged functional group have high water solubilities, whereas FTOH, FOSA, and N-EtFOSE have much lower water-solubilities (Ahrens, 2011) since their hydrophilic functional heads are uncharged (Table 1). As surfactants, PFAAs are likely to aggregate at the interface between octanol and water, and log K_{OW} values which are an indicator of compound hydrophobicity, are therefore difficult to determine experimentally (Tolls et al., 1994; Tolls and Sijm, 1995). When interpreting log K_{OW} values obtained through modelling

this surfactant behaviour should be kept in mind. Sorption studies of long chain PFASs in sediment revealed that log K_{OC} values increased with increasing fluorocarbon chain length (Higgins and Luthy, 2006; Ahrens et al., 2010).

3. PFASs in drinking water

In comparison to occurrence surveys in surface and groundwater, fewer finished drinking water occurrence studies are available. Table SI-2 in the supporting information (SI) lists studies that have reported occurrence of PFOS and PFOA in treated drinking/tap water worldwide. A summary of global PFOA/PFOS occurrence data is presented in Fig. 1. Although instances of μg/L concentrations of PFASs in drinking water have been reported (Emmett et al., 2006; Skutlarek et al., 2006; Minnesota Department of Health, 2008), detected concentrations are typically in the lower ng/L range provided that there
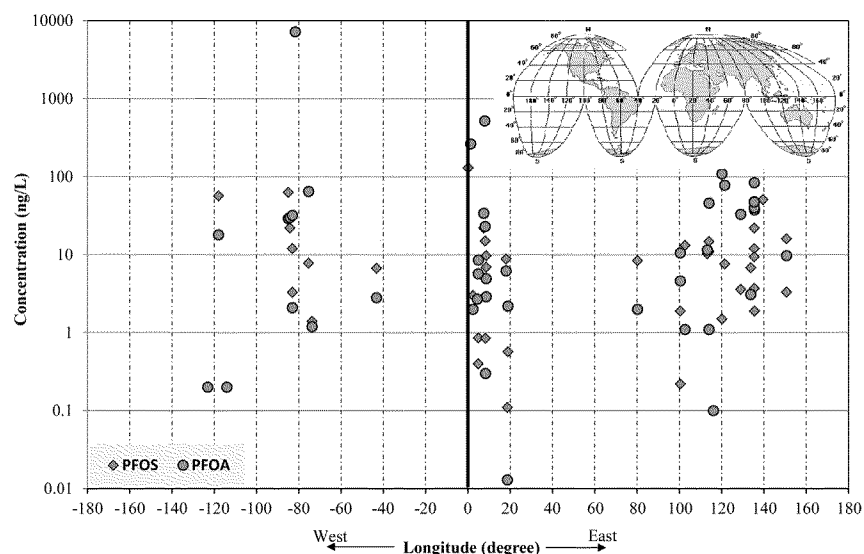
Fig. 1 – Reported global concentration of PFOS/PFOA in drinking water by longitude (locations are approximate and were obtained using Google Earth). Detailed data and study references can be found in the supporting information Table SI-2.

is no obvious PFAS point source close to a drinking water treatment plant intake. Drinking water occurrence studies have typically targeted PFOS and PFOA, and as a result these two are the most commonly detected compounds. Hence, this discussion focuses primarily on PFOS and PFOA. However, other compounds including PFBA, PFPA, PFHxA, PFHpA, PFNA, PFUnDA, PFHxS, and FOSA have also been detected in drinking water (e.g. Wilhelm et al., 2010; Ahrens, 2011; Ullah et al., 2011). For instance, PFBA was detected at a mean concentration of ~2000 ng/L in treated water entering the City of Oakdale, Minnesota, distribution system which is adjacent to the 3M Cottage Grove PFAS manufacturing facility (Minnesota Department of Health, 2008). Some recent European studies have detected the shorter chain replacement PFASs such as PFBA, PFBS, and PFHxA in drinking water at concentrations even higher than PFOA and/or PFOS at some locations (Ullah et al., 2011; Eschauzier et al., 2012) indicating the change in production and usage patterns. PFBA and PFBS detected at average concentrations of 30 ng/L and 20 ng/L, respectively, were the highest detected PFASs in finished water collected from a treatment plant in Amsterdam (Eschauzier et al., 2012). Branched isomers of PFOS and PFOA have also been detected in drinking water (Eschauzier et al., 2012).

High concentrations of PFOA has been detected in the Little Hocking community adjacent to the DuPont fluoropolymer manufacturing facility in Washington, West Virginia (Fig. 1, near 80). PFOA was detected in the distribution system at an average concentration of 4800 ng/L (range 487–10,100 ng/L) (Paustenbach et al., 2007) and in private drinking water wells in surrounding communities at a mean concentration of 200 ng/L (Hoffman et al., 2011). Data from the Little Hocking Water Association indicate that PFOA was present at μg/L levels in raw water prior to GAC treatment and varied from 2400 ng/L to 8500 ng/L in the period from October, 2007 to April, 2010 (Little Hocking Water Association, 2010). High concentrations of PFOA in drinking water (500–640 ng/L) were also reported in the Arnsberg-Neheim, Sauerland area,

Germany in 2006 (Skutlarek et al., 2006). Subsequent investigation identified an agricultural area, where organic soil conditioners mixed with industrial waste were applied, as the contamination point source. A study by Quinones and Snyder (2009) monitoring seven US drinking water utilities demonstrated that the occurrence and concentration of PFASs are more likely to be higher in the finished waters of treatment plants whose raw water sources are impacted by wastewater treatment plants than those that are pristine or less impacted by wastewater discharge. PFASs in finished water have also been detected in the UK, China, Canada, India, Japan, Poland, and Sweden. Typical concentrations in drinking water in different countries are quite comparable (<50 ng/L PFOS; <100 ng/L PFOA) (Fig. 1), except for the point source contamination scenarios in Germany, the United States, and the United Kingdom. PFASs have also been detected in bottled water (Rostkowski et al., 2008; Kunacheva et al., 2010) and in tap water-based beverages including coffee and cola (Eschauzier et al., 2013).

While dietary intake is likely one of the important exposure routes to PFOA and PFOS (Haug et al., 2011), in the previously described cases in Little Hocking, US, and Arnsberg, Germany, drinking water was found to be the major exposure route (Emmett et al., 2006; Hölzer et al., 2008). Concentrations of PFOA in blood plasma of inhabitants of Arnsberg, Germany were 4.5–8.3 times higher compared to a nearby reference population where PFASs were not detected in drinking water. The higher blood plasma PFOA level in Arnsberg residents was found be to clearly associated with consumption of tap water and PFOA concentrations were higher in residents who consumed more tap water at home (Hölzer et al., 2008). The concentration of PFOA in Little Hocking water was about 7-fold higher compared to Arnsberg and the mean serum level PFOA concentration of the population from Little Hocking was 16–18 fold higher compared to that of Arnsberg residents. In an effort to reduce the concentration of PFOA in drinking water, granular activated carbon (GAC) filters were installed in

both cases. Follow-up studies noted that GAC adsorption decreased the levels of PFOA in treated water to below their limits of detection (Hölzer et al., 2009; Bartell et al., 2010), however, GAC needed frequent replacement or regeneration to maintain this level of PFOA removal (see Section 4.3). In both cases blood serum level PFAS concentrations decreased by as much as 28% over the year following the installation of the GAC filters.

Table 3 — Reported full-scale drinking water treatment plant PFASs removal data.

Water source	Treatment	Raw/influent [ng/L] (frequency/ season/month)		Finished/tap water [ng/L] (frequency/ season/month)	Percent removal[d] (%)	Reference
PFOS						
Groundwater	DBF, UV, Cl$_2$	10.0 (100%)		9.4 (100%)	6	Quinones and
Surface water	O$_3$, COA/FLOC, DBF, Cl$_2$	1.4 (67%)		1.4 (64%)	0	Snyder (2009)
Surface water	PAC, CHLM, DBF	1.7 (50%)		1.9 (43%)	12	
Surface water	Cl$_2$, COA/FLOC, DBF, UV	22 (100%)		22 (100%)	0	
Planned potable indirect reuse facility	MF/RO, UV/H$_2$O$_2$, SAT	41 (100%)		ND	100	
Planned potable indirect reuse facility	Cl$_2$, DL, SAT	29 (100%)		57 (100%)	97	
River water	RSF, O$_3$, GAC, Cl$_2$	1.0 (Summer)		0.93 (Summer)	7	Takagi
River water	RSF, O$_3$, GAC, Cl$_2$	0.87 (Summer)		2.8 (Summer)	222	et al. (2008)
		3.2 (Winter)		1.6 (Winter)	50	
Lake water	RSF, GAC, Cl$_2$	4.6 (Summer)		0.16 (Summer)	97	
		4.5 (Winter)		<0.1 (Winter)	>98	
River, lake, subsoil and groundwater (data from seven plants)	RSF, Cl$_2$	0.56—22 (Sum) 0.54—4.2 (Win)		0.45—22 (Sum) 0.37—4.5 (Win)	20—0 31 to 7	
River water	Membranes (no further information), Cl$_2$	0.37 (Summer) 0.26 (Winter)		0.29 (Summer) 0.20 (Winter)	22 23	
Lake water	SSF, Cl$_2$	2.7 (Summer) 1.8 (Winter)		2.3 (Summer) 1.9 (Winter)	15 6	
River water	COA/FLOC/SED, SF, O$_3$, GAC, Cl$_2$	1.3 (Summer) 3.3 (Winter)		3.7 (Summer) 1.3 (Winter)	185 60	Takagi et al. (2011)
River water	COA/FLOC/SED, SF, O$_3$, GAC, Cl$_2$	1.6 (Summer) 3.3 (Winter)		2.3 (Summer) 1.7 (Winter)	44 48	
River water	COA/FLOC/SED, SF, O$_3$, GAC, Cl$_2$	1.2 (Summer) 2.8 (Winter)		1.6 (Summer) 1.9 (Winter)	33 32	
River water	SED, O$_3$, GAC, Cl$_2$, SF	1.4 (Summer) 3.3 (Winter)		2.2 (Summer) 2.0 (Winter)	57 39	
Lake water	COA/FLOC/SED, SF, GAC (reactivated), Cl$_2$	4.4 (Summer) 4.1 (Winter)		<0.5 (Summer) <0.5 (Winter)	>89 >88	
Groundwater	UF, Cl$_2$	16		16	0	Atkinson
Groundwater	GAC (not in operation), super chlorination and dechlorination	135		130	3	et al. (2008)
Groundwater	GAC (2 parallel GAC trains each having 6 beds; contactors are mature and act as biological contactors; not been regenerated for some years), Cl$_2$	59[a] 29[a] 38[a]	42[b]	45 (post GAC 42 ng/L)	7	
Ground and surface water (60:40)	SSF, O$_3$, GAC (6 beds- no regeneration for several years), Cl$_2$ using NaOCl	21[a] 28[a] 20[a]	20.6[c]	25	21	
River water	COA/FLOC/SED, O$_3$, GAC, RSF	5.3 (Aug) 5.8 (Oct)		9.4 (Aug) 6.4 (Oct)	77 (Aug) 10 (Oct)	Shivakoti et al. (2010)
River water	COA/FLOC/SED, O$_3$, GAC, RSF	5.8 (Aug) 8.8 (Oct)		3.9 (Aug) 4.2 (Oct)	33 (Aug) 53 (Oct)	
Treated wastewater	De-nitrification, pre O$_3$, COA/FLOC/SED, DAFF, O$_3$, GAC (acts as biological contactors), O$_3$	2.2 (Oct) 3.7 (Nov) 3.6 (Nov)		<LOR (0.3 ng/L) (Oct) 0.6 (Nov) 0.7 (Nov)	100 (Oct) 84 (Nov) 81 (Nov)	Thompson et al. (2011b)

Table 3 — (continued)					
Water source	Treatment	Raw/influent [ng/L] (frequency/ season/month)	Finished/tap water [ng/L] (frequency/ season/month)	Percent removal[d] (%)	Reference
River water	COA/FLOC/SED, RSF, Cl_2	5.02	0.73	85	Kunacheva et al. (2010)
Treated wastewater	Clarifier/lamellar settler ($FeCl_3$ & $(NH_4)_2SO_4$, NaOCl addition), UF, RO, UV/H_2O_2, Stabilization/ disinfection (addition of lime, CO_2, NaOCl)	38 39 23	<LOR (0.5 ng/L) ND <LOR (0.2 ng/L)	100 100 100	Thompson et al. (2011b)
River water	COA/FLOC, RSF, O_3, GAC, SSF	8.2	<0.23	<97	Eschauzier et al. (2012)
River water	Cl_2, COA/FLOC, RSF, O_3, GAC	116	33	69[e]	Flores
River water	Cl_2, COA/FLOC, RSF, O_3, GAC, UF, RO	86	13	86[e]	et al. (2013)
PFOA					
Groundwater	DBF, UV, Cl_2	11 (100%)	11 (100%)	0	Quinones and
Surface water	O_3, COA/FLOC, DBF, Cl_2	5.6 (3%)	<MRL (5 ng/L)	~0[f]	Snyder (2009)
Surface water	PAC, CHLM, DBF	9 (17%)	<MRL (5 ng/L)	~0[f]	
Surface water	Cl_2, COA/FLOC, DBF, UV	31 (100%)	30 (100%)	3	
Planned potable indirect reuse facility	MF/RO, UV/H_2O_2, SAT	15 (100%)	ND	100	
Planned potable indirect reuse facility	Cl_2, DL, SAT	25 (100%)	18 (100%)	28	
River water	COA/FLOC/SED, SF, O_3, GAC, Cl_2	15 (Summer) 24 (Winter)	48 (Summer) 24 (Winter)	220 0.0	Takagi et al. (2011)
River water	COA/FLOC/SED, SF, O_3, GAC, Cl_2	33 (Summer) 26 (Winter)	42 (Summer) 25 (Winter)	27 4	
River water	COA/FLOC/SED, SF, O_3, GAC, Cl_2	10 (Summer) 19 (Winter)	22 (Summer) 20 (Winter)	120 5	
River water	SED, O_3, GAC, Cl_2, SF	26 (Summer) 26 (Winter)	36 (Summer) 31 (Winter)	38 19	
Lake water	COA/FLOC/SED, SF, GAC (reactivated), Cl_2	42 (Summer) 42 (Winter)	6.5 (Summer) 9.2 (Winter)	85 78	
River water	COA/FLOC/SED, O_3, GAC, RSF	32.0 (Aug) 31.6 (Oct)	24.0 (Aug) 47.5 (Oct)	25 (Aug) 50 (Oct)	Shivakoti et al. (2010)
River water	COA/FLOC/SED, O_3, GAC, RSF	12.0 (Aug) 33.2 (Oct)	12.0 (Aug) 46.3 (Oct)	0 (Aug) 39 (Oct)	
River water	RSF, O_3, GAC, Cl_2	25 (Summer)	32 (Summer)	28	Takagi
River water	RSF, O_3, GAC, Cl_2	64 (Winter)	84 (Winter)	31	et al. (2008)
River water	RSF, O_3, GAC, Cl_2	19 (Summer) 58 (Winter)	15 (Summer) 35 (Winter)	21 40	
Lake water	RSF, GAC, Cl_2	67 (Summer) 92 (Winter)	6.9 (Summer) 4.1 (Winter)	90 92	
River, lake, subsoil and groundwater (data from seven plants)	RSF, Cl_2	8.4–58 (Sum) 8.4–42 (Win)	6.9–40 (Sum) 7.1–31 (Win)	18–31 15–26	
River water	Membranes (no further information), Cl_2	5.2 (Summer) 7.4 (Winter)	2.3 (Summer) 5.0 (Winter)	56 32	
Lake water	SSF, Cl_2	28 (Summer) 32 (Winter)	21 (Summer) 19 (Winter)	25 41	
Treated wastewater	De-nitrification, pre O_3, COA/FLOC/SED, DAFF, O_3, GAC (acts as biological contactors), O_3	6.1 (Oct) 16 (Nov) 13.6 (Nov)	7.6 (Oct) 10.9 (Nov) 12.1 (Nov)	24 (Oct) 32 (Nov) 11 (Nov)	Thompson et al. (2011b)
Treated wastewater	Clarifier/lamellar settler ($FeCl_3$ & $(NH_4)_2SO_4$, NaOCl addition), UF, RO, UV + H_2O_2, Stabilization/ disinfection (addition of lime, CO_2, NaOCl)	22 27 15	<LOR (0.7 ng/L) <LOR (0.7 ng/L) <LOR (0.9 ng/L)	100 100 100	

Table 3 — (continued)

Water source	Treatment	Raw/influent [ng/L] (frequency/season/month)	Finished/tap water [ng/L] (frequency/season/month)	Percent removal[d] (%)	Reference
River water	Cl₂, COA/FLOC, RSF, O₃, GAC	21	13	52[e]	Flores et al. (2013)
River water	Cl₂, COA/FLOC, RSF, O₃, GAC, UF, RO	6.9	3.0	89[e]	
Groundwater	UF, Cl₂	25	66	164	Atkinson et al. (2008)
Groundwater	Cl₂	155	183	18	
Groundwater	IX, nitrate removal, Cl₂, phosphate dosing	55	59	7	
Groundwater	air stripping, Cl₂	182	263	45	
Groundwater	GAC (2 parallel GAC trains each having 6 beds; contactors are mature and act as biological contactors; not been regenerated for some years), Cl₂	46[a] 44[b] / 45[a] / 41[a]	66	50	
Ground and surface water (60:40)	SSF, O₃, GAC (6 beds- no regeneration for several years), Cl₂ using NaOCl	48[a] 55.4[c] / 66[a] / 31[a]	71	28	
Groundwater	Cl₂ using NaOCl	105[a] 111.5[b] / 118[a]	125	12	
River water	Cl₂, COA/FLOC, RSF, O₃, GAC	21	13	52[e]	Flores et al. (2013)
River water	Cl₂, COA/FLOC, RSF, O₃, GAC, UF, RO	6.9	3.0	89[e]	
River water	COA/FLOC/SED, RSF, Cl₂	9.57	1.79	81	Kunacheva et al. (2010)
River water	COA/FOC, RSF, O₃, GAC, SSF	4.4	5.1	16	Eschauzier et al. (2012)
PFHxA					
Groundwater	DBF, UV, Cl₂	1.5 (67%)	1.4 (83%)	7	Quinones and Snyder (2009)
Surface water	O₃, COA/FLOC, DBF, Cl₂	1.2 (30%)	1.2 (39%)	0	
Surface water	PAC, CAM, DBF	1.1 (33%)	1.1 (14%)	0	
Surface water	Cl₂, COA/FLOC, DBF, UV	29 (100%)	23 (100%)	21	
Planned potable indirect reuse facility	Cl₂, DL, SAT	14 (100%)	1.9 (100%)	86	
Planned potable indirect reuse facility	MF/RO, UV/H₂O₂, SAT	7.9 (100%)	ND	100	
Treated wastewater	De-nitrification, pre O₃, COA/FLOC/SED, DAFF, O₃, GAC (acts as biological contactors), O₃	6.5 (Oct) / 4.4 (Nov) / 4.4 (Nov)	5.2 (Oct) / 6.0 (Nov) / 6.5 (Nov)	20 (Oct) / 36 (Nov) / 48 (Nov)	Thompson et al. (2011b)
Treated wastewater	Clarifier/lamellar settler (FeCl₃ & (NH₄)₂SO₄, NaOCl addition), UF, RO, UV/H₂O₂, Stabilization/disinfection (addition of lime, CO₂, NaOCl)	13 / 14 / 11	ND / ND / ND	100 / 100 / 100	
PFHxS					
Groundwater	DBF, UV, Cl₂	2.1 (83%)	2.2 (100%)	5	Quinones and Snyder (2009)
Surface water	PAC, CAM, DBF	2.5 (33%)	1.4 (43%)	44	
Surface water	Cl₂, COA/FLOC, DBF, UV	12 (100%)	12 (100%)	0	
Planned potable indirect reuse facility	Cl₂, DL, SAT	5.1 (100%)	6.1 (100%)	20	
Planned potable indirect reuse facility	MF/RO, UV/H₂O₂, SAT	9.3 (100%)	ND	100	
Treated wastewater	De-nitrification, pre O₃, COA/FLOC/SED, DAFF, O₃, GAC (acts as biological contactors), O₃	1.5 (Oct) / 2.3 (Nov) / 2.1 (Nov)	1.1 (Oct) / 1.5 (Nov) / 2.0 (Nov)	27 (Oct) / 35 (Nov) / 5 (Nov)	Thompson et al. (2011b)
Treated wastewater	Clarifier/lamellar settler (FeCl₃ & (NH₄)₂SO₄, NaOCl addition), UF, RO, UV/H₂O₂, Stabilization/disinfection (addition of lime, CO₂, NaOCl)	36 / 28 / 12	<LOR (0.4 ng/L) / <LOR (0.1 ng/L) / <LOR (0.3 ng/L)	100 / 100 / 100	
River water	COA/FOC, RSF, O₃, GAC, SSF	2.0	0.6	70	Eschauzier et al. (2012)
PFBA					
River water	COA/FOC, RSF, O₃, GAC, SSF	33	30	9.1	Eschauzier et al. (2012)

Table 3 — (continued)

Water source	Treatment	Raw/influent [ng/L] (frequency/ season/month)	Finished/tap water [ng/L] (frequency/ season/month)	Percent removal[d] (%)	Reference
PFBS					
Treated wastewater	De-nitrification, pre O_3, COA/FLOC/SED, DAFF, O_3, GAC (acts as biological contactors), O_3	ND (Oct) ND (Nov) ND (Nov)	1.7 (Oct) 0.8 (Nov) 1.3 (Nov)	– – –	Thompson et al. (2011b)
Treated wastewater	Clarifier/lamellar settler (FeCl$_3$ & (NH$_4$)$_2$SO$_4$, NaOCl addition), UF, RO, UVH$_2$O$_2$, Stabilization/ disinfection (addition of lime, CO$_2$, NaOCl)	6.4 4.8 2.4	<LOR (0.1 ng/L) ND ND	100 100 100	
River water	COA/FOC, RSF, O_3, GAC, SSF	35	20	43	Eschauzier et al. (2012)

AC — activated carbon, CHLM — chloramination, Cl$_2$ — Chlorination, COA/FLOC/SED — coagulation/flocculation/sedimentation, DAFF — dissolved air flotation and sand filtration, DBF — deep bed filtration, DL — dilution, UV — medium pressure ultraviolet, GAC — granular activated carbon, MF/RO — microfiltration/reverse osmosis, NaOCl — sodium hypochlorite, O_3 — ozonation, PAC — powder activated carbon, RSF — rapid sand filtration, SSF — slow sand filtration, SAT — soil aquifer treatment.
ND — not detected; LOR — limit of reporting.
[a] Concentration of compound in intake from groundwater borehole (session 1).
[b] Calculation: average concentration of groundwater borehole intakes.
[c] Calculation: 0.4   surface water concentration + 0.6   average concentration of 3 groundwater boreholes.
[d] % removal estimated using the formula (1   C/C$_0$)   100% and rounded; where C$_0$ is the raw/influent water concentration and C is the effluent/ tap water concentration (when ND or <LOR, a value of zero was assigned).
[e] Overall % removal reported by Flores et al. (2013).
[f] PFOA was detected at concentrations below the method reporting limit (MRL) in both influent and effluent samples but could not be quantified. For each utility only one influent sample contained PFOA in concentrations slightly above the MRL. Hence, it is likely that no significant removal took place.

# 4. PFAS removal during drinking water treatment

Treatment efficiency is expected to vary widely across classes of perfluorinated compounds due to differences in their physical–chemical properties. Only a few studies focussing on PFAS removal during full-scale drinking water treatment were located which is not surprising considering the relatively recent emergence of this issue and the fact that they are disbursed throughout the scientific literature. These are, however, sufficient in number to be able to make some preliminary observations.

PFAS plant surveys quickly demonstrated that conventional treatment processes were unable to substantially remove PFASs. For example, Tabe et al. (2010) reported that PFOA and PFOS were detected in more than 90% of treated water samples collected from drinking water treatment plants in the Detroit River watershed (highest occurrence frequency among 51 micro-contaminants monitored). To further illustrate this observation, a list of selected PFAAs (PFOA, PFOS, PFHxA, and PFHxS) reported in both raw and finished water at full-scale plants has been compiled (Table 3). This table lists only studies that provided some details on the treatment schemes employed. Raw water or influent concentrations ranged from 0.4 to 182 ng/L and are similar to what is typically observed in surface water surveys in general. Observed influent and effluent concentrations at the majority of the listed plants are similar indicating minimal removal of PFASs through treatment. Fig. 2 clearly illustrates, that with the exception of nanofiltration (NF) and reverse osmosis (RO),

water treatment technologies used at the treatment plants, including ozonation and advanced oxidation, failed to achieve appreciable PFAS removals. In fact, in several instances, detected concentrations of PFOA and PFOS in finished water were higher than in raw water prior to treatment (Fig. 2 and Table 3). While analytical error at these extremely low analyte concentrations may be partially responsible, breakdown of certain precursor compounds to PFOS and PFOA during treatment may also be possible (Takagi et al., 2008; Shivakoti et al., 2010). Other potential sources for higher finished water concentrations include leaching from Teflon -coated treatment equipment components (Tabe et al., 2010) and desorption from GAC filters that had been in service for long periods without reactivation (Takagi et al., 2011). Shorter chain PFASs concentrations, in particular, may be higher after treatment as a result of desorption from GAC due to competition for active sorption sites with longer chain PFASs (Eschauzier et al., 2012) or natural organic matter (NOM) constituents.

4.1. Conventional coagulation, flocculation, sedimentation, and filtration

The extremely low concentrations of PFASs, together with their high hydrophilicity, make them unlikely candidates for removal by conventional coagulation/flocculation/sedimentation processes. In fact, no differences in PFAS concentrations were found between plant influent and sedimentation unit effluent samples collected from two drinking water treatment plants in Kansai, Japan (Shivakoti et al., 2010).
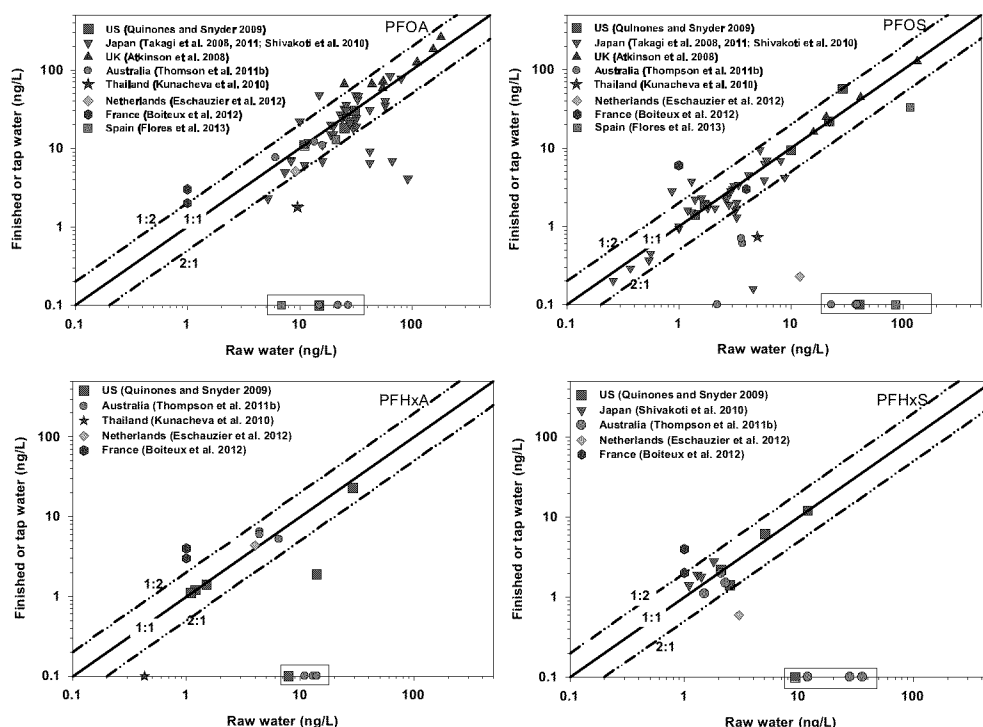
Fig. 2 – Reported finished and raw drinking water **concentration of selected PFASs at various full scale plants. A value of 0.1 ng/L was assigned when PFAS concentrations were either below the limit of detection (< LOD) or limit of reporting (< LOR) or not detected (ND). Boxed data points denote data from plants that use NF/RO membranes indicating high PFAS removals at those plants.**

Similarly, PFAS concentrations in samples collected from five full-scale plants in Osaka, Japan following coagulation and sedimentation, and sand filtration preceded by sedimentation, indicated that essentially no removal took place through either combination of unit processes (Takagi et al., 2011). Similarly, no removals by conventional coagulation treatment were reported by Thompson et al. (2011b) and Eschauzier et al. (2012). This is also consistent with a bench-scale coagulation study investigating PFOA and PFOS removal, which found removals of less than 35% under a variety of conditions tested (Xiao et al., 2012b).

Eschauzier et al. (2010) based on their study of surface, ground and drinking waters commented that both rapid- and slow-sand filtration are unlikely to be effective for PFAS removal. This is supported with observations made by Takagi et al. (2008, 2011), Shivakoti et al. (2010), Eschauzier et al. (2012), and Flores et al. (2013). However, Kunacheva et al. (2010) observed that rapid sand filters achieved high removals of PFOA (85%) and PFOS (86%) in the particulate phase but low removals for the aqueous phase. The latter being consistent with the other reports.

4.2. Oxidation processes

Fluorine is the most electronegative element and as such resists oxidation to retain its electrons. Being the most powerful inorganic oxidant (redox potential $E^0 = 3.06$ V) (Beltrán, 2004), it is thermodynamically unfavourable to oxidize fluorine. The presence of functional groups with high electron density such as double bonds, activated aromatic systems, and amino groups generally increase the reactivity of a compound with ozone (O_3) ($E^0 = 2.07$ V), while the presence of electron withdrawing groups (e.g. $-Cl$, $-NO_2$, $-COOH$) lowers their reactivity (von Gunten, 2003). PFAAs do not contain aromatic bonds or phenolic structures (Table 1). Thus, the presence of the strong C–F bond together with the electron withdrawing functional groups $-COOH$ and $-SO_3H$ in the structures of PFCAs and PFSAs, respectively, indicates that these compounds will likely be resistant to oxidation, even by molecular ozone and hydroxyl radicals. Hydroxyl radicals ($\cdot$OH) ($E^0 = 2.8$ V), the primary oxidant in advanced oxidation processes (AOPs), generally withdraw H-atoms from saturated organics to form water thus PFAAs due to perfluorination (i.e. replacement of all hydrogen by fluorine) are also unlikely candidates for oxidation by AOPs. Szajdzinska-Pietek and Gebicki (2000) found that PFOA was not very reactive with $\cdot$OH, and they estimated the upper limit of the second order reaction rate constants for $\cdot$OH with PFOA to be 3×10^7 M^{-1} S^{-1}, which is quite low for reactions with $\cdot$OH. For example, the estimated upper limit of the reaction rate is at least two orders of magnitude lower than the average reaction rate between $\cdot$OH and sodium octanoate (5.6×10^9 M^{-1} S^{-1}), the corresponding unfluorinated hydrocarbon of PFOA (Szajdzinska-Pietek and Gebicki, 2000), and thus PFAAs are not likely to be easily degraded by AOPs. Based on the low reactivity of PFAAs with ozone and in AOPs it is expected that chlorine-based oxidation processes, due to their lower redox potentials ($E^0 = 1.36–1.50$ V), will also very likely not oxidize PFASs under typical drinking water treatment conditions.

Limited full-scale treatment plant surveys conducted to-date confirm these theoretical considerations in that chlorine and ozone-based oxidation processes, at typical water treatment plant doses and contact times, were not effective for the removal of PFASs (Atkinson et al., 2008; Quinones and Snyder, 2009; Takagi et al., 2011). PFASs have been shown to be resistant to chlorination or chloramination even when combined with other unit processes such as coagulation/flocculation/sedimentation, powdered activated carbon (PAC), deep bed filtration, and UV irradiation (Quinones and Snyder, 2009). Inefficacy of chlorine-based oxidants for PFAS removal during drinking water treatment has also been reported by Atkinson et al. (2008) and Takagi et al. (2011). Ozone-based oxidation processes have been reported to fail to transform PFAAs (Takagi et al., 2008, 2011; Tabe et al., 2010; Shivakoti et al., 2010; Thompson et al., 2011b; Eschauzier et al., 2012; Flores et al., 2013). At a full-scale water reclamation plant in Australia, even multiple stages of ozonation with doses as high as 5 mg/L with 15 min contact time failed to achieve PFAS removal (Thompson et al., 2011b). Ozone doses and contact times as high as 0.87 mg/L and 120 min, respectively, were not effective for PFOA and PFOS removal (Takagi et al., 2011) [ozone residuals not available for either study above]. PFOS and PFOA can be formed from the degradation of precursor compounds such as FASAs, FASEs and FTOHs. These precursors are mostly polyfluorinated compounds thereby containing C–H bonds which may be oxidizable. Thus, if ozone or advanced oxidation processes (AOPs) were able to oxidize polyfluorinated precursors present in the raw water, the concentration of terminal compounds such as PFOS or PFOA may actually increase in finished water. Further studies are needed to resolve this.

4.3. Granular activated carbon adsorption

GAC is widely used in drinking water treatment plants for reducing the concentrations of synthetic organic contaminants, taste and odour compounds, and sometimes natural organic matter (NOM). GAC has been used to treat PFASs in a few full-scale installations (Atkinson et al., 2008; Minnesota Department of Health, 2008; Hölzer et al., 2009; Little Hocking Water Association, 2010; Takagi et al., 2011; Eschauzier et al., 2012; Flores et al., 2013). GAC filters when new, or in use for less than nine months, were found to achieve 69–100% removal of ng/L level PFOS and PFOA at five treatment plants in Osaka, Japan (Takagi et al., 2011).

Sorption capacity of virgin activated carbon used in one of the plants studied by Takagi et al. (2011) was estimated to be about 520 ng/g considering flow, GAC volume, and concentration of PFASs in GAC influent (empty bed contact time, hydraulic loading, and GAC type were not specified). Although under very different conditions, Hansen et al. (2010) using Langmuir isotherms estimated a maximum PFOA sorption capacity in a similar range with 1100 ng/g for GAC in contaminated groundwater.

Eschauzier et al. (2012) observed that only the GAC filters (Norit ROW 0.8 Supra), and not the preceding coagulation, rapid sand filtration, and ozonation steps, were effective in removing PFASs in a treatment plant in Amsterdam, Netherlands. While GAC alone effectively removed PFNA,

PFOS and PFHxS, it only partially removed PFOA (~50%) and failed to remove shorter chain PFASs such as PFBA, PFBS, PFPA, PFHxA and PFHpA (Eschauzier et al., 2012). Flores et al. (2013) reported partial removal of both PFOA (41%) and PFOS (63%) by GAC adsorbers (containing Filtrasorb 400 , Norit ROW 0.8 and Norit 1240 EN) when these compounds were present at low ng/L levels in the raw water at a Spanish drinking water treatment plant. When looking into isomer-specific behaviour of PFOA and PFOS during GAC treatment it was found that branched isomers were less sorbable to GAC compared to linear isomers (Eschauzier et al., 2012).

GAC filters (containing Calgon Carbon Filtrasorb 100) were installed in a water treatment plant in Arnsberg, Germany to treat PFAS-contaminated water in July 2006 (Hölzer et al., 2009). PFOA was not detected in water samples collected during the next two months (Fig. 3A). In late August, 2006, however, re-appearance of PFOA was observed and its level eventually exceeded the precautionary value of 100 ng/L in early December, 2006 at which point the GAC was reactivated (Hölzer et al., 2009). The Little Hocking Water Association, Ohio, US also reported frequent replacement (~3 months) of GAC (Calgon Carbon Filtrasorb 600) to achieve PFOA removal from drinking water at albeit elevated influent concentrations (1900–8500 ng/L) (Fig. 3B) (Little Hocking Water Association, 2010). Takagi et al. (2011) also observed that GAC when not reactivated for longer periods (>1 year), was unable to effectively remove PFOA and PFOS. They further observed that once activated, GAC lasted for about 130 days until the re-appearance of PFOA in the GAC filtered water. A reduction in the service life of GAC filters used for PFAS removal due to NOM preloading was also noticed by Eschauzier et al. (2012). The City of Oakdale, Minnesota started using GAC filters in October 2006 at a newly constructed pilot plant to remove PFASs from groundwater using two GAC filters in series, each filter containing 20,000 pounds of GAC (Minnesota Department of Health, 2008). PFBA was the first compound to be detected between the first and second set of GAC filters at the plant after only six weeks of operation while breakthrough of PFOA and PFOS were observed after 286 days and 550 days, respectively (Minnesota Department of Health, 2008; Kolstad, 2010). Kolstad (2010) reported that the Oakdale plant, by replacing GAC based on PFOA breakthrough, was able to treat 1.9 billion gallons of water over a period of 23 months which amounted to a GAC replacement cost of about $0.12 per 1000 gallons of water. Early breakthrough of PFBA is also consistent with Eschauzier et al. (2012) who did not observe removal of PFBA. Decreased log K_{OC} values of PFASs with decreasing carbon chain length (Higgins and Luthy, 2006; Ahrens et al., 2010) indicate lower sorption potential of shorter chain PFASs compared to their longer chain counterparts. This may explain the observed earlier breakthrough of PFBA.

Two important adsorption phenomena that arise during treatment of natural water due to the presence of dissolved organic carbon (DOC) are competitive adsorption and preloading or fouling of GAC. It is likely that both direct competition and in particular preloading phenomena are responsible for the observed early breakthrough of PFASs during GAC filtration at full-scale treatment plants. Slow sorption kinetics of PFASs onto GAC may also contribute to early breakthrough (Yu et al., 2009a). Failure to reactivate or replace GAC likely
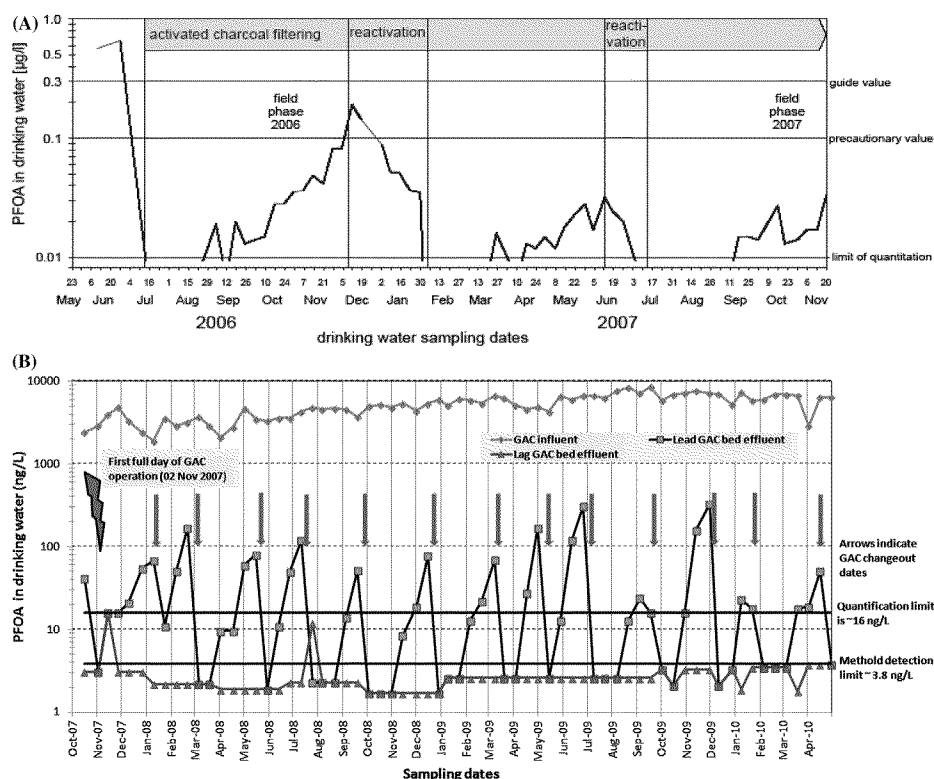
Fig. 3 – (A) PFOA-concentration in drinking water in Arnsberg, Germany between May 2006 and April 2008 indicating frequent need for GAC filter reactivation for PFOA removal (Hölzer et al., 2009; reprinted with permission from the publisher). Please note, PFOA concentration is reported in this figure in μg/L. Calgon F-100 ™ was used as the GAC at the treatment plant. GAC info collected via personal communication with the corresponding author. (B) PFOA-concentration in raw and finished drinking water at Little Hocking, West Virginia, USA. Calgon F-600 ™ GAC used at the treatment plant. Data collected from Little Hocking Water Association (2010) and by personal communication with Mr. Bob Griffin, General Manager, Little Hocking Water Association.

explains why Atkinson et al. (2008) did not see any removal of PFASs at water treatment sites where GAC filters were in place but had not been regenerated for years. Reactivating carbon 2–3 times per year has been suggested to achieve and maintain good removal of PFASs (Takagi et al., 2011) but this strategy has considerable implications in terms of cost and operations. Taking into account the low health-based guideline values being suggested for PFOA and PFOS (Table 2), GAC applications specifically targeting PFASs need to be carefully designed and optimized to reduce the frequency of activated carbon regeneration. Once in place it may require enhanced monitoring to assess performance and to determine timing of the regeneration.

PFAS isotherms and kinetic parameters in ultrapure water at environmentally relevant concentrations may provide an initial basis for evaluating the suitability of a particular type of carbon for PFAS treatment. Studies in natural water will be useful to assess pre-loading and direct competition effects. Previously, Yu et al. (2009b) observed that GAC preloaded for 16 weeks had about 2–10% of its capacity remaining for the hydrophilic and ionic compound naproxen. PFAAs also have hydrophilic and ionic mioeties and their adsorption when present at trace concentrations may as well be severely impacted by NOM preloading. Yu et al. (2009b) using other non PFAS trace contaminants also demonstrated that isotherms

generated at high concentrations, if used to extrapolate capacity at very low target contaminant concentrations, may result in overestimation of GAC removal capacity. Thus for isotherm studies it is important to employ concentrations which are similar to those encountered in natural water.

Reported data from full-scale treatment applications demonstrate that PFAS breakthrough may occur relatively early in GAC adsorbers, but the actual breakthrough time is compound- and water-specific. Therefore, pilot-scale studies are likely needed to optimally design filters or contactors thereby providing the basis for balancing capital investment in terms of filter design, carbon cost, and frequency of regeneration. Pilot-scale studies in natural water at environmentally relevant PFAS concentrations may assist in obtaining more accurate assessments of GAC adsorption capacity that may be encountered under real water treatment scenarios.

4.4. Powdered activated carbon adsorption

Powdered activated carbon (PAC) has also been studied for PFAS removal (Qu et al., 2009; Yu et al., 2009a; Hansen et al., 2010; Dudley, 2012) but only at bench-scale. Dudley (2012) found that thermally activated wood-based PAC was more efficient in removing PFASs when compared to coconut,

lignite, and bituminous PAC. In buffered ultrapure water (pH 7.0), at an initial PFAS concentration of 500 ng/L, thermally activated wood-based PAC at a dose of 15 mg/L achieved >70% removal of eight target PFAS within 15 min of contact time. However, less than 40% removal of PFPeA was observed, with no removal for the shorter chain PFBA, confirming the negative effect of decreasing hydrophobicity with decreased carbon chain length on adsorption. Similar to GAC, PFAS adsorption on PAC is also negatively affected by the presence of NOM. The same thermally activated wood-based PAC at the same dose in North Carolina reservoir water in the presence of 4.5 mg/L of TOC achieved a maximum of only 55% removal for PFDA (C10) and PFOS. The study concluded that significant removal of PFBA, PFPeA, PFHxA, PFHpA, and PFBS from drinking water may not be achieved at practical PAC dosages (Dudley, 2012).

Experiments with PFOA and PFOS not surprisingly indicate that PFAS adsorption kinetics are much faster for PAC compared to GAC. About 168 h and 4 h were required to reach equilibrium for GAC and PAC, respectively, for both compounds (Yu et al., 2009a). Higher PFAS removal using PAC (60–90%) as opposed to GAC (20–40%) in short duration adsorption tests (10 min) were observed by Hansen et al. (2010) at trace concentration levels and in the presence of NOM. Thus, PAC likely adsorbs PFASs faster than GAC due to its smaller particle size resulting in higher surface area for the same volume of carbon, shorter internal diffusion distances, and additional available surface functional groups (Yu et al., 2009a; Hansen et al., 2010). Also, the poorer performance of GAC relative to PAC may be attributable to the rigidity of the CF_2 backbone which may not energetically favour sorption into the inner pores of GAC (Hansen et al., 2010). PFASs have been detected in water throughout the year and hence, GAC adsorbers may be a better long term solution if PFAS is the contaminant of concern. PAC may be a more appropriate choice for removing PFASs in situations that require a prompt short-term response (e.g. spills).

4.5. Biodegradation

PFAAs will likely not be biodegraded under typical drinking water treatment conditions. Although reductive defluorination appears to be thermodynamically favourable and releases enough energy for microbes to thrive, the compounds do not seem to be commonly used as a carbon source (Parsons et al., 2008). Meesters and Schroder (2004) reported complete removal of both PFOS and PFOA from wastewater samples under anaerobic conditions in a lab-scale closed-loop bioreactor, however, biodegradation was not observed under aerobic conditions. In-plant biological drinking water treatment processes operate almost exclusively under aerobic conditions thereby not creating conditions favourable for reductive defluorination. Microbial metabolization of FTOHs and the FTOH-based products, FASAs, FASEs as well as other PFAA precursor compounds has been reported to occur during wastewater treatment (under aerobic conditions) and in the environment (Wang et al., 2005a,b; Rhoads et al., 2008; Martin et al., 2010) and may eventually lead to formation of PFAAs (e.g. PFOS, PFOA). Degradation of precursors to PFAAs during

drinking water treatment remains to be systematically investigated.

4.6. High pressure membranes

Wastewater reclamation and reuse programs, desalination, and the demand for high quality drinking water are some of the driving forces behind the increasingly growing application of high pressure nanofiltration (NF) and reverse osmosis (RO) membrane processes. The viability of high pressure membrane applications is improving with advances in energy efficiency, operating efficiency, lowered costs, and the ability of membranes to tackle a wide range of water contaminants. In general, high pressure membrane processes are not widely used for the treatment of drinking water other than in the case of localized specific contaminants, softening, and desalination. PFASs, due to their presence at considerably higher concentrations in wastewater compared to surface water, are of concern for drinking water utilities that are employing or are planning to adopt water reclamation or reuse programs.

Low pressure membranes such as microfiltration membranes (MF) alone will not be able to retain PFASs as the effective diameter of these molecules are smaller (~1 nm) compared to MF pore sizes which are in the range of ~100 nm (Tsai et al., 2010). Available bench-scale studies involving high pressure membranes indicate that membrane pore size/molecular weight cut-off (MWCO) probably plays the most important role with respect to rejection of PFASs by NF/RO (Tang et al., 2006, 2007; Steinle-Darling and Reinhard, 2008; Lipp et al., 2010; Appleman et al., 2013). High removals of charged PFASs with a size of 300 Da or greater can be expected. For charge neutral PFASs such as FOSA, rejection may vary and can be substantially lower (Steinle-Darling and Reinhard, 2008; Steinle-Darling et al., 2010). While size is probably the dominant factor, solute–membrane interactions which will depend on factors such as charge, hydrophobicity, and dipole moment are also expected to be significant if the solute molecular weight is close to or smaller than the MWCO of the membrane. Adsorption onto membrane surfaces (Kwon et al., 2012) and back diffusion can also play important roles in the rejection of PFASs. Membrane fouling layers may hinder back diffusion of the retained PFAS molecules which eventually facilitates transport of the retained solutes across the membrane thereby decreasing net rejection (Steinle-Darling and Reinhard, 2008). However, contrasting results showing better performance of fouled membranes in rejecting PFAAs were reported by Appleman et al. (2013). This is not surprising as others have observed an increase in rejection for pharmaceuticals for fouled membranes filtering water from different sources (Comerton et al., 2009). Under certain conditions Comerton et al. (2009) also reported a decrease in rejection.

It was observed in a study involving two Australian water reclamation plants that the one with an RO unit preceded by a UF unit and followed by an advanced oxidation process (AOP) (UV + H_2O_2) unit achieved almost complete removal (not detected or below detection limit) of PFASs (Thompson et al., 2011b). A slight decrease in the concentration of some PFASs following the UF unit was attributed to the removal of suspended and colloidal particles with which PFASs may have been associated. Much higher concentration of PFASs in the

RO concentrate compared to feed water corroborates that PFASs were primarily removed by the RO unit (Thompson et al., 2011b). In contrast, no decrease in PFOA and other shorter PFAS concentrations was observed in the finished water of the other plant that had three ozonation stages located at different points in the treatment train and a biological activated carbon filtration stage (in addition to conventional coagulation). Quinones and Snyder (2009) in their survey of seven US drinking water utilities observed that PFASs were only removed at a utility whose treatment included an RO unit. Complete removal (99%) of PFOA and PFOS following RO membrane treatment has also been reported by Flores et al. (2013). Data collected from these studies strongly suggest that high pressure membranes are capable of substantial PFAS removal (Table 3 and Fig. 2). This is consistent with bench-scale studies conducted with ng to μg/L concentrations of PFASs found in surface water (Loi-Brügger et al., 2008; Steinle-Darling and Reinhard, 2008; Lipp et al., 2010; Appleman et al., 2013).

Bench-scale studies have for the most part been conducted in water matrices lacking DOC. Rejection mechanisms can be affected by the presence of DOC in water and hence, future studies are needed to elucidate PFAS behaviour during membrane filtration in the presence of DOC. An issue inherent to contaminant removal by membrane processes is the disposal of the PFAS-enriched concentrate which will have to be carefully considered.

4.7. Resin treatment

PFAAs, being anionic at ambient water pH values, would be expected to be amenable to removal by anion exchange. Hence, this discussion focuses predominantly on strong base anion exchange resins. Electrostatic interactions as well as adsorption via hydrophobic interactions are the two primary mechanisms proposed for removal with ion exchange resins. Transport to binding sites may also play a role. The pH of typical drinking waters (6–9) is not expected to have any significant effect on removal by ion exchange due to the ionization of PFOA and PFOS. Important resin characteristics that may affect removal include functional groups, polymer matrix, and porosity (Deng et al., 2010). It is unclear from existing studies which mechanism prevails and if it varies among PFAAs. Thus, the term 'uptake' when used in this section indicates binding to the resins by both electrostatic and hydrophobic interactions.

Only one study reported full-scale demonstration of PFAS removal by ion exchange from raw water used for drinking water production. Purolite FerrIX A33 , a strong base, porous anion exchange resin impregnated with iron oxide was used at a New Jersey DWTP for arsenic removal. It was observed that at low level (ng/L) PFAS influent concentrations appreciable removal of longer chain PFCAs (54% for PFHpA and 76% for PFOA) and high removal of PFSAs (83%, >97% and >90% for PFBS, PFHxS and PFOS, respectively) (Dickenson et al., 2012) was achieved. However, the resin failed to remove shorter chain PFCAs (PFBA, PFPeA and PFHxA). Magnetic ion exchange (MIEX) which is predominantly used for DOC removal, was also reported to be ineffective (<10%) for the removal of PFASs at a plant in Alabama (Dickenson et al., 2012).

In addition to binding by electrostatic and hydrophobic interactions, transport to binding sites may also play a role in the uptake of PFASs. Acrylic resins are more hydrophilic than styrenic resins. Hence, acrylic resins may achieve better removal of hydrophilic PFASs as they facilitate transport to the acrylic resin pores. This hypothesis is supported by the results of Deng et al. (2010), who, when studying PFOS removal during wastewater treatment with ion exchange resins, observed that polyacrylic resins, regardless of resin porosity and functional groups, had faster uptake rates and higher equilibrium capacities than did polystyrene resins. Similar trends have also been observed by Lampert et al. (2007) and Dudley (2012). Dudley (2012) reported that although macroporous polyacrylic strong base anion resin had faster uptake kinetics, the resin exhibited lower uptake capacity compared to both the gel and macroporous types of polystyrenic strong base anion resins used in their study.

Study results of Deng et al. (2010) further indicate that macroporous resins are expected to exhibit better uptake compared to gel resins due to easier accessibility to resin exchange sites. Hydrophilicity and the open structure of macroporous resins probably facilitate uptake of PFAAs by inducing faster diffusion into the anion exchange sites. Dudley (2012), however, observed that uptake kinetics for macroporous polystyrenic and gel type polystyrenic resins were similar.

Compared to activated carbon, a significantly improved removal of shorter chain PFASs has been reported with strong base anion resins. Polystyrenic strong base anion resin achieved >90% removal of PFBA and PFPeA at 'doses' of 5 and 10 mL/L in natural water. The author hypothesized (but could not confirm) that NOM potentially alters the resins in a way that facilitates PFAS uptake (Dudley, 2012).

Non-ion exchange resins have also been tested at benchscale for removal of PFASs (Senevirathna et al., 2011; Xiao et al., 2012c; Chularueangaksorn et al., 2013). Findings of Xiao et al. (2012c) show that moderately polar non-ionic Amberlite XAD-7HP performed better than the non-polar Amberlite XAD-2 resin. The authors also indicated regeneration did not significantly affect performance of the XAD-7HP resin. Chularueangaksorn et al. (2013) however, observed that anionic resins had higher sorption capacity for PFOA compared to non-ionic resins.

Regardless of some of the contrasting trends observed during the studies conducted to-date, it is evident that resin treatment has the potential to be a promising technology for the removal of PFASs from water. However, resin studies to-date were mostly conducted in the absence of DOC and results may be different in its presence. Thus, further investigations are warranted before recommending ion exchange for PFAS. It is also important to note that when selecting an ion exchange resin, regeneration issues can be as important as the removal capacities of the resin. The presence of other competing anions (e.g. SO_4^{2-}, NO_3^-) should also be considered as they may also affect uptake capacity of resins. Another consideration is the potential for breakthrough and a subsequent contaminant spike (dumping) into the treated water as the resin approaches exhaustion. Moreover, it may be challenging to elucidate uptake mechanisms and trends as typically the exact structure and nature of ion exchange sites

for various commercially available resins are proprietary in nature.

5. Knowledge gaps and research needs

The current knowledge gap with regard to an adequate physico-chemical property database of PFASs creates a challenge for the assessment of the fate of PFASs. Limited information is available about isomeric profiles for PFCAs and PFSAs. Since isomers are also likely to be present in the aquatic environment (Houde et al., 2008), and considering the recent observation that linear isomers are preferentially sorbed onto GAC compared to their branched counterparts (Eschauzier et al., 2012), the behaviour and fate of isomers of various PFASs during drinking water treatment needs to be investigated.

The presence of precursor compounds may play a role as they may convert to terminal products such as PFOA and PFOS during drinking water treatment and may therefore lead to increased concentrations in finished water. As such, removal and degradation studies of PFAS precursors are also warranted.

Most studies to-date have focused on PFOA and PFOS, however, as new PFASs, for example shorter chain PFASs are introduced (Renner, 2006) it is likely that those compounds will eventually become significant contributors to total PFAS levels in drinking water. Data on PFAS occurrence in finished drinking water are still limited and even sparse for some of the more recently detected PFASs. Thus human exposure to these compounds via water is still poorly understood. Future studies and regulatory considerations need to consider that PFASs found in the aquatic environment may eventually be detected in drinking water. Limited but available data suggest that those shorter chain PFASs are also more challenging to treat.

Although efforts are underway to regulate the production of some PFASs (USEPA, 2009), they will remain on the market, at least in the near future, and continue to be detected in the environment. Thus, it is becoming increasingly evident that both understanding of the fate of PFASs during drinking water treatment as well as optimization of existing treatment schemes will be necessary if there are societal or regulatory pressures to remove these compounds.

Finally, better coordination among regulatory bodies in different jurisdictions in terms of understanding, character-izing, and minimizing the risk of exposure to PFASs via drinking water is desirable. Such initiatives would minimize the wide variations in prevailing emergency regulatory guidelines and will help utilities set realistic treatment goals if this becomes necessary.

6. Conclusions

This article identifies the limitations of present day drinking water treatment technologies and potential advantages of currently less-exploited technologies (ion exchange and high pressure membrane filtration). This compilation of available full-scale drinking water removal surveys/studies of per-fluoroalkyl and polyfluoroalkyl substances (PFASs), along with select bench-scale studies suggests that:

Conventional coagulation, flocculation, and sedimentation cannot achieve substantial removal (<20%) of PFASs nor can rapid granular media filtration.

Free chlorine at residuals commonly employed for disin-fection or distribution system residual maintenance is ineffective for PFAS removal.

Oxidation and advanced oxidation processes, under typical drinking water treatment plant conditions, will not oxidize most PFASs. Some oxidation of FTOHs and FASAs may be possible; however, they may simply be oxidized to other PFASs.

UV irradiation at commonly utilized disinfection doses and at the higher doses used for contaminant removal is also ineffective.

GAC may be useful for removing PFASs from drinking water. Longer chain PFASs will sorb better onto sorbents compared to the shorter chain compounds. However, short chain PFASs such as PFBA and PFBS may pass through or reach breakthrough very quickly. The efficiency of GAC is compromised in the presence of NOM and frequent carbon reactivation may be necessary. Future studies should consider the elucidation of the effects of preloading and direct competition in natural water on the PFAS removal efficiency by activated carbon adsorption.

Biodegradation of most PFASs in aerobic GAC contactors or in other forms of biofiltration used under current drinking water treatment conditions is unlikely.

Ion exchange/non-ion exchange resins, while not commonplace in drinking water treatment facilities, may be useful for removing PFASs. Additional data is needed to understand the effect of resin type and water matrix (competing anions and NOM). Resin regeneration and disposal of brine needs to be taken into consideration.

NF/RO membranes will achieve high rejection of most PFASs. However, lower molecular weight PFASs (such as PFBA, PFPeA), and the neutral FOSA may be less well rejected by some loose NF membranes. Data on rejection following long term operation of membranes and in the presence of NOM are not available. Disposal of concentrate, which will contain elevated concentrations of PFASs, will need to be addressed.

Appendix A. Supplementary material

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.watres.2013.10.045.

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